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# Dynamical and chemical contributions to variability in Microwave Limb Sounder Arctic stratospheric column ozone

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**Abstract.** Analyses of column ozone above 100 hPa (Col100) derived from Upper Atmosphere Research Satellite Microwave Limb Sounder (MLS) data in February/March 1992-1998 show that about half of the interannual variability in Col100 in the Arctic polar vortex in late winter results from interannual variability in chemical loss. A majority of the remainder results from interannual variability in day-to-day dynamical motions including adiabatic warming/cooling and poleward advection of underlying upper tropospheric subtropical air on short timescales, rather than from variations in descent rates and large-scale transport over the winters. The morphology of Col100 from MLS remains very similar to that in the dynamical models even in the years with most chemical ozone loss. The amount and character of day-to-day variability in dynamical models closely follows that in MLS Col100. Although the morphology of and day-to-day variability in Arctic column ozone are controlled by dynamical processes, chemical ozone loss was a major factor in producing both the low values of and the large interannual variability in Arctic column ozone observed during the 1990s.

## 1. Introduction

Many studies demonstrate a strong relationship between Arctic column ozone amounts and day-to-day variations in dynamical processes [e.g., *Petzoldt*, 1999; *Orsolini et al.*, 2001; *Hood et al.*, 2001, and references therein], and several have shown strong dynamical contributions to trends in column ozone in northern hemisphere (NH) midlatitudes [e.g., *Fusco and Salby*, 1999; *Hood et al.*, 1999, and references therein]. NH March average 63-90°N column ozone from the Total Ozone Mapping Spectrometer (TOMS) was lower in the 1990s than in the previous decade [e.g., *Newman et al.*, 1997; *Andersen and Knudsen*, 2002]. Calling this a "trend" may be an overstatement because there is little evidence to support a trend in either the 1980s or the 1990s when taken separately [e.g., *Andersen and Knudsen*, 2002] and other studies have reported no evidence of a trend within the 1990s [e.g., *Chipperfield and Jones*, 1999]. The observed decrease in column ozone in the 1990s is often presented as evidence of chemical ozone loss, and substantial chemical loss related to chlorine and bromine destruction processes was indeed observed in the Arctic lower stratospheric vortex during the 1990s [e.g., *Solomon*, 1999; *WMO*, 1999]. However, a decrease in column ozone during this period might be expected even in the absence of chemical loss since the lower stratosphere was also unusually cold during most of the 1990s [e.g., *Pawson et al.*, 1998; *Pawson and Naujokat*, 1999], and lower column ozone results from a decrease in lower stratospheric temperatures via dynamical as well as chemical processes [e.g., *Fioletov et al.*, 1997, and references therein]. Not only is low column ozone associated with low temperature in day-to-day dynamical variations [e.g,. *Petzoldt et al.*, 1994; *Petzoldt*, 1999; *Hood et al.*, 2001; *Weber et al.*, 2002, and references therein], but also less diabatic descent occurs in colder winters, resulting in less ozone being transported to the lower stratosphere where it contributes most to column values. There is large interannual variability in column ozone, and much higher ozone was seen in recent warm winters than during the unusually cold winters in the 1990s [e.g., *Andersen and Knudsen*, 2002]. Thus, variability, both in-

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terannual and intraseasonal, in high-latitude column ozone arises from a combination of dynamical and chemical effects.

*Chipperfield and Jones* [1999] showed chemical transport model simulations for the 1990s in which the majority of interannual variability in high-latitude column ozone resulted from dynamical variations; their modeled 63-90°N March-mean column showed ∼8 DU variability in chemical loss out of a total interannual variability of ∼21 DU in 1992 through 1998. They attributed the dynamical contribution to variations in diabatic descent and hence in large-scale, seasonal transport during the winter. *Andersen and Knudsen* [2002] used vortex column loss estimates based on observations (from different methods and datasets in different years) with vortex area estimates to show that vortex chemical loss could account for up to ∼75% of the decrease in 63-90°N column ozone between the 1980s and the 1990s, and that interannual variability in the 1990s in the absence of chemical loss would be expected to be about half of that observed. Although *Andersen and Knudsen* [2002] presented their results as being in contrast to those of *Chipperfield and Jones* [1999], the inconsistency appears to be largely a matter of exact magnitude when comparing results related to interannual variability (the only results that are comparable between the two studies, as *Chipperfield and Jones* [1999] could not quantify values before the 1990s), with about 38 and 52% of the variability attributed to chemical loss by *Chipperfield and Jones* [1999] and *Andersen and Knudsen* [2002], respectively.

Both the *Chipperfield and Jones* [1999] and the *Andersen and Knudsen* [2002] studies are characterized by large uncertainties. Many studies [e.g., *Davies et al.*, 2002; *Manney et al.*, 2002b, and references therein] show large uncertainties in the effect of modeled transport on polar processing studies, and additional uncertainties arise in modeling the chemical processes. *Andersen and Knudsen* [2002] analyzed estimates of chemical loss having large individual uncertainties [e.g., *Harris et al.*, 2002], and used estimates from different methods and datasets in different years, raising the possibility of interannual variability in biases between the estimates; additional uncertainties arise from the assumptions used to convert vortex chemical loss to 63-90°N loss. Uncertainties in the observed estimates shown by *Newman et al.* [1997] and updated in *Andersen and Knudsen* [2002] are also substantial, since they are taken from several different TOMS instruments (the value for the 1996 NH winter is in fact from a Solar Backscatter Ultraviolet (SBUV) rather than a TOMS instrument), and in addition do not represent a complete 63-90°N March average, since TOMS observes only to ∼70°N at the beginning of March, reaching near the pole only in late March. Thus significant uncertainties remain regarding how much of the observed variability in column ozone may be attributed to chemical versus dynamical processes.

Here, we focus on the polar vortex region, where wintertime chemical loss resulting from heterogeneous reactions on polar stratospheric clouds (PSCs) occurs. From the Upper Atmosphere Research Satellite (UARS) Microwave Limb Sounder (MLS) we have available a consistent set of ozone data during seven consecutive NH winters in the 1990s from the same instrument, extending to 80°N during all observing periods. *Manney et al.* [2002a] have estimated chemical ozone loss, including vortex loss in column above 100 hPa, from this dataset. We use their Lagrangian transport (LT) model results to estimate the relative effects of chemical processing versus all dynamical processes, and another, simpler, model to isolate the effects of short timescale processes such as adiabatic warming/cooling and quasi-isentropic transport. With the MLS data and these models, we examine the relative roles of dynamical and chemical processes in producing both day-to-day and interannual variability in stratospheric column ozone in the Arctic vortex region.

## 2. Column Ozone from MLS and Dynamical Models

MLS gathered ozone data during the 1991-1992 through 1997-1998 Arctic winters; *Livesey et al.* [2002] and *Manney et al.* [2002a] provide more detailed information on MLS observational coverage. The MLS instrument switches between viewing ∼34°S to 80°N and ∼80°S to 34°N approximately every 36 days, giving coverage of high northern latitudes in separate early and late winter periods. The MLS viewing orientation switch occurred a few days earlier in each successive winter observed by MLS, so the late winter MLS north-viewing period covered from mid-February to late March in 1992, but by 1998 it covered late January to late February. Observations were daily in 1992 and 1993, and nearly so in 1994. In later years, MLS observations were limited by spacecraft power constraints and by the need to conserve the lifetime of the MLS antenna scanning mechanism; thus observations in these years were typically no more frequent than every other day (two out of every three days in late February 1996).

Column ozone above 100 hPa (Col100) is calculated from version 5 MLS data [*Livesey et al.*, 2002] by integrating the retrieved abundances over the MLS retrieval grid (6 points per decade change in pressure). Tropopause pressures range from ∼120 to 300 hPa in Arctic winter, so Col100 represents most, but not all, of the stratospheric column. Comparisons of Col100 with TOMS total column (not shown) indicate similar morphology and evolution in daily maps, and very

similar patterns of variability in Col100 and TOMS column averaged in the lower stratospheric vortex (where comparable averages can be computed, since TOMS does not observe up to 80°N until ∼10 March). *Froidevaux et al.* [1994] also showed similar time variations in zonal mean MLS Col100 and TOMS data. Spatial correlation coefficients (r) between Arctic late winter MLS Col100 and TOMS total column are typically $r \gtrsim 0.80$ (a conservative calculation using an empirical orthogonal functions analysis to estimate the degrees of freedom for a Students-t test [*Bretherton et al.*, 1999] gives a 95% significance level of $r \approx 0.60$). MLS did not observe low enough altitudes to examine total column ozone, but the region observed by MLS does cover the levels where most of the vortex chemical ozone loss (∼85 to 100%, e.g., *Goutail et al.* [1999, 2001]) takes place. Although they do not continuously cover the Arctic, during the periods that are observed MLS data have the advantages of providing coverage of polar night and of seven consecutive winters in the 1990s that include the years with largest observed Arctic ozone loss. In contrast to results from solar occultation or ground-based observations (such as those analyzed by *Andersen and Knudsen* [2002]), the relatively complete hemispheric coverage of MLS on each viewing day allows us to examine the morphology of Col100 in relation to the vortex, temperatures, and other meteorological variables, facilitating better understanding of the origins of variability seen in vortex averages.

In addition, since we have full three-dimensional (3D) ozone fields from MLS, we can use MLS data as input for transport/dynamical models. *Manney et al.* [2002a] used Arctic winter MLS ozone observations with an LT model to estimate ozone loss on isentropic (potential temperature, θ) surfaces from 385-840 K. Here, we calculate Col100 from three different sources: from the MLS data, from a simple dynamical model based on reconstructing column ozone from potential vorticity (PV)/θ-space "climatologies", and from the LT model runs described by *Manney et al.* [2002a]. These calculations are used to investigate the relative roles of chemical and dynamical processes in producing variability in Arctic Col100 during the seven late winters observed by MLS; the simple dynamical model is used to explore the importance to interannual variability of short timescale dynamical variations, and to estimate dynamical variability in regions/times not observed by MLS.

Figure 1 shows MLS Col100 averaged over 14-28 February, for 1992-1998. This period was chosen because it is the latest winter period during which MLS data are available in each of the seven years. This period includes three MLS observing days in 1995, two in 1998, and more in the other years, up to 15 in 1993. The lower stratospheric vortex position (on the 465-K potential temperature surface) and cold regions (at 46 hPa) are also shown. Large interannual variability is seen in Col100, temperature, and the relative positions of the low column ozone/cold regions and the vortex. Low Col100 is co-located with low temperature and is often poorly spatially correlated with the vortex region, as has been seen in many previous studies [e.g., *Petzoldt et al.*, 1994; *Manney et al.*, 1996a, b; *Hood et al.*, 2001, and references therein], and as is discussed further in section 3. Localized, very low Col100 regions are seen in 1993, 1998, and particularly 1996. During these periods, there was a high, cold tropopause (not shown) under the low Col100 region, and low values of upper tropospheric PV (typical of the subtropics) were advected into high latitudes under the low Col100 area; these are conditions characteristic of the formation of localized extreme ozone minima (miniholes) [e.g., *Hood et al.*, 2001; *Allen and Nakamura*, 2002; *Weber et al.*, 2002, and references therein].

*Orsolini et al.* [2001] used a reverse trajectory model initialized with MLS ozone data mapped as a function of equivalent latitude (EqL)[1] and potential temperature (θ) to model high-resolution ozone profiles throughout the 1997-1998 winter at the ALOMAR station in the high Arctic. These profiles were used to reconstruct column ozone values, and reproduced most observed day-to-day variations at ALOMAR during the 1997-1998 winter. *Allen and Nakamura* [2002] reconstructed column ozone during a deep minihole event by combining data from several solar occultation instruments using a "tracer" EqL coordinate (i.e., EqL calculated from a passive tracer field generated in a high-resolution transport model driven by analyzed winds) to produce 3D hemispheric ozone fields and obtained good agreement with observed values. Here we use an idealized dynamical model (the "EqL" model) similar to those described above: We reconstruct 3D ozone distributions by mapping an EqL/θ ozone "climatology" into physical space using daily PV and temperature fields (several climatologies have been used as noted below); Col100 is calculated from these reconstructed distributions in the same manner as from MLS data. We thus assume that the large-scale, winterlong transport processes are the same in each winter; since the initial EqL/θ field on a given day is the same in each year, the only variations that affect this model are the shape of the PV field and the pressure of the isentropes on that day. Hence, Col100 from the EqL model only reflects variations in the morphology and evolution of the PV field (including, possibly, poleward advection of low-PV, low-ozone air during any minihole events), and the adiabatic motions of the isentropes.

[1] The latitude that would enclose the same area between it and the pole as a given potential vorticity (PV) contour, e.g., *Butchart and Remsberg* [1986]

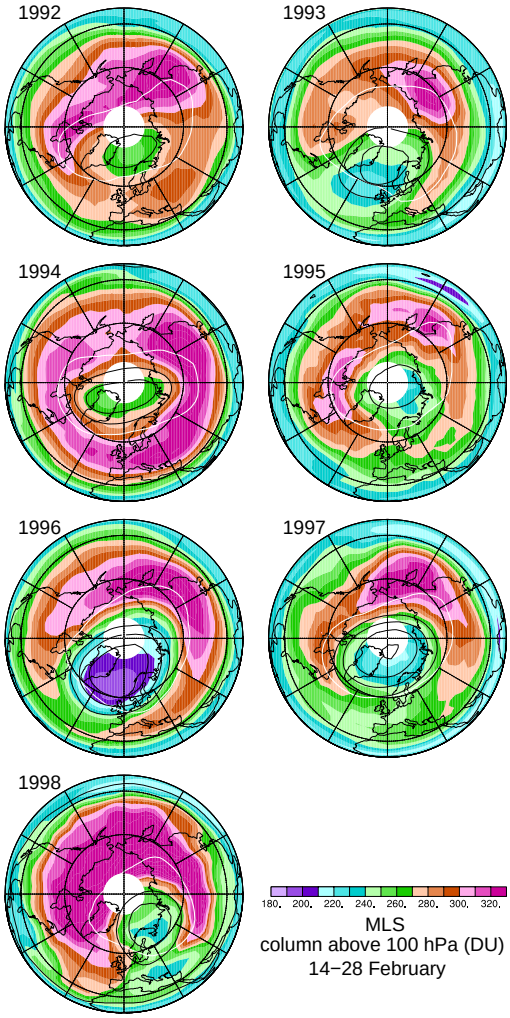


**Figure 1.** 14-28 February average MLS Col100 (DU) maps for 1992-1998, with 465-K PV in the vortex edge region (white contour), and 205, 200, 195, and 190 K 46-hPa temperatures (black, outer to inner, 205 K contour appears on all maps) from UK Met Office data overlaid. Projection is orthographic, with 0°E at the bottom and 90°E to the right; dashed circles are at 30° and 90°N.

Figure 2 shows 14-28 February averages constructed using a climatology based on EqL/θ averages of all MLS data in 10-day bins throughout the winter [*Manney et al.*, 2002a]. A reconstruction based on MLS data from the warm 1997-1998 winter (when there was little ozone loss) produces slightly more interannual variability, with lows similar to those shown here but maxima that are higher than those in Figure 2 (not shown). The considerable interannual differences seen here and their similarity to those in the MLS Col100 fields (Figure 1) indicate that adiabatic motion of the isentropes and the evolution of the PV field are significant factors contributing to interannual variability in Col100. Spatial correlation coefficients for the area north of 40°N calculated for all daily fields from MLS and the EqL model during the MLS late winter observing periods are $0.76 \leq r \leq 0.94$ (as noted above, the 95% significance level is $r \approx 0.6$), with largest values in 1996. 1996 was the year in which MLS saw most chemical ozone loss [*Manney et al.*, 2002a]; the 1996 late winter period was also one during which the cold and low column ozone regions were located along the vortex edge (Figure 1), and minihole activity was strong and frequent [e.g., *Manney et al.*, 1996b; *Weber et al.*, 2002]. Under these conditions, MLS column ozone would be expected to become less well spatially correlated with that from a purely dynamical model as chemical loss increased, since chemical loss is rapidly (especially in a highly distorted vortex) spread throughout the vortex region and would thus not be well-correlated with the Col100 field. The fact that the spatial correlation between MLS and EqL model fields is particularly high during 1996 indicates that dynamical processes play a dominating role, especially during minihole events, in determining the morphology of column ozone.

Col100 was calculated from the LT model results as described by *Manney et al.* [2002a]. Briefly, LT ozone was interpolated to the 100-10 hPa pressure surfaces, and relaxed to MLS observations over 22-10 hPa (since the LT model does not include a parameterization of low-latitude ozone production in the mid-stratosphere, it is not expected to be accurate at these levels, which contribute very little to Col100, and in which there is no evidence of vortex chemical loss due to heterogeneous processing). Missing LT ozone at 100 hPa was filled, if possible, by extrapolation, and gaps of one or two gridpoints were filled with a weighted average of surrounding data. If good data were not then present at all levels from 100-22 hPa, Col100 at that location was treated as missing data. *Manney et al.* [2002a] used the LT model calculations initialized with MLS ozone at the beginning of each late winter observing period (referred to hereinafter as "LTL" runs); the differences between these calculations and the MLS observations thus reflect the chemical loss that oc-

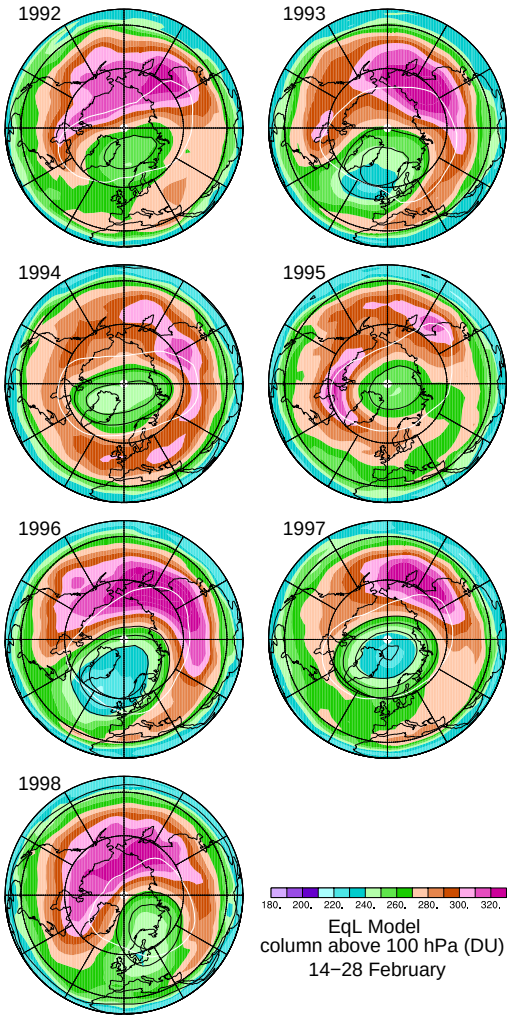


**Figure 2.** 14-28 February Col100 (DU) from EqL model (see text), in 1992-1998. Layout is as in Figure 1.

curred after the beginning of those observing periods. The 14-28 February period we focus on in much of the following was near the beginning of the LTL runs in 1992 and 1993, and thus results from these runs (not shown) strongly resemble the MLS observations during that period. Using "extended" LT calculations (referred to hereinafter as "LTE" runs), wherein the results of LT model runs for mid-winter were used to reinitialize for the late winter period, *Manney et al.* [2002a] also produced estimates of cumulative ozone loss observed by MLS. In most years there was no evidence of chemical loss before the beginning of the LTE runs (ranging from mid-January in 1992 to early December in 1998), so the LTE runs combined with the MLS data provide a good estimate of the total chemical loss up to their ending date (perhaps a slight underestimate in the 1991-1992 and 1993-1994 winters, when there is evidence of a small amount of ozone loss in early winter before the beginning of the first part of the LTE runs).

14-28 February averages from the LTE runs (Figure 3) show the LT model predictions of column ozone in the absence of chemical loss. While these show values substantially higher than observed by MLS (Figure 1) in most years, considerable interannual variability is still evident, and the morphology of the LTE Col100 fields is very similar to that of the MLS fields. Spatial correlation coefficients for daily MLS and LTE fields throughout all the LTE runs are $0.60 \leq r \leq 0.93$, with the vast majority of them above 0.70. In 1993 and 1996 (the two years in which MLS observed most chemical ozone loss), there is a slight decrease in the correlation coefficients (from $\sim$0.85 to $\sim$0.75 in 1993 and $\sim$0.90 to $\sim$0.80 in 1996) over the MLS late winter observing period, suggesting that as more chemical loss occurs and is distributed throughout the vortex, the observed column ozone may become more correlated with the vortex and thus less well-correlated with the dynamically calculated column ozone. However, even in these years, the morphologies of the Col100 fields from MLS and the LTE model runs resemble each other much more strongly than either one resembles the shape of the vortex.

## 3. Dynamical and Chemical Variability in MLS Column Ozone

Figure 4 shows maximum, average and minimum Col100 from MLS and the dynamical models, for the 1992-1998 MLS late winter observing periods, in the lower stratospheric vortex region (defined by the 1.2 $\times 10^{-4} s^{-1}$ scaled PV contour at 465 K, white overlay in Figures 1-3). Since chemical ozone loss via heterogeneous processing on PSCs in the lower stratosphere takes place almost entirely within the vortex [e.g., *WMO*, 1999; *Manney et al.*, 2002a, and ref-

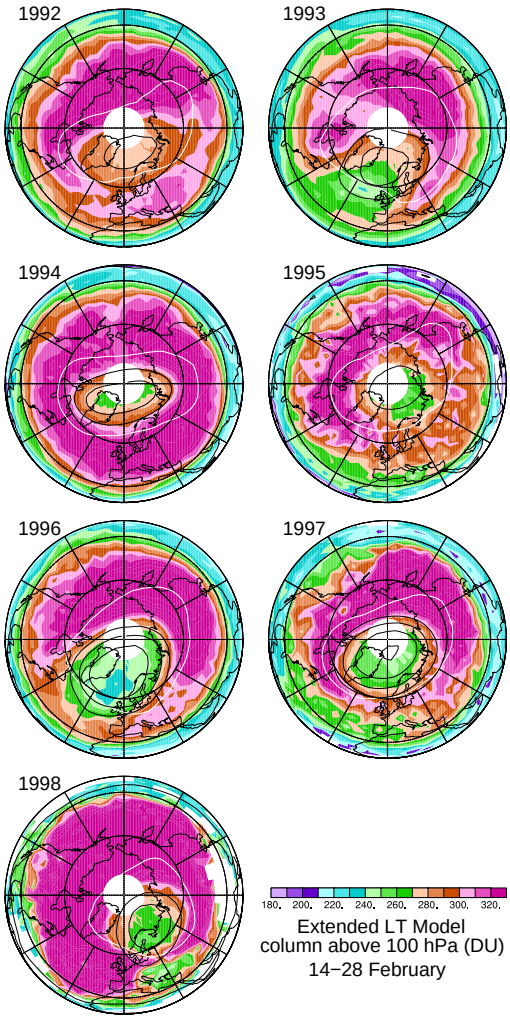


**Figure 3.** 14-28 February Col100 (DU) from LTE runs (see text), in 1992-1998. Layout is as in Figure 1.

erences therein], averaging over the vortex region encompasses the area where both dynamical and chemical processes are expected to be contributing significantly to column ozone changes.

The precision of individual version 5 MLS Col100 values is ∼7 DU; since the averages are over hundreds of measurements, their precision is very high. Since the climatological fields used in the EqL model come from averages of many MLS observations, uncertainties in the EqL model values arise primarily from uncertainties in the PV fields used in the reconstruction. While it is difficult to quantify those uncertainties, a rough idea can be obtained by using PV from different meteorological datasets in the reconstruction and for the vortex area definition. Differences between the results shown in Figure 4 and those calculated using PV from the National Centers for Environmental Prediction (NCEP)/Climate Prediction Center (CPC) analyses are up to ∼20 DU on individual days in the extrema and ∼10 DU in the average. In all cases, however, the amounts of interannual and day-to-day variability are within 1-3 DU of each other. Daily extrema in the LTL and LTE runs are quite uncertain, especially in the latter part of the periods shown, since they represent a single grid-point value from a transport calculation 20-40 days long; however, they give a suggestion of the type of behavior expected. The variability in and evolution of daily LTL and LTE vortex averages are expected to be good to within 10-20% [*Manney et al.*, 2002a]. Minimum LTL and LTE values for 14–21 February 1996 are particularly suspect because that period was cold enough that the lowest LT model isentrope, 385 K, was above 100 hPa at a few points, which were filled by extrapolation/interpolation of surrounding values; the LTE run for 1998 is also not thought to be as reliable, as the mid-winter LT run it was reinitialized from was longer than the period for which LT calculations are expected to be accurate [*Manney et al.*, 2002a]. In addition, *Manney et al.* [2002a] showed evidence, although not conclusive, for a slight overestimate of the modeled increase in ozone in the 1992 late winter (but not in the other years).

Comparison of the range of values in individual years between each of the models and MLS indicates that day-to-day variability is comparable in MLS and the dynamical models. During 1992 through 1994, when MLS data were nearly daily, the temporal correlation coefficients between MLS and the dynamical model vortex averages are significant at the 99% confidence level (the degrees of freedom for each series are estimated using a measure of the autoregressive property [*Livezey and Chen*, 1983] and are used to calculate the confidence level [*Freund*, 1971]), indicating a close correlation of day-to-day changes. Deep minihole events occurred around 20 February and 3 March 1996 [e.g., *Manney*

*et al.*, 1996b; *Weber et al.*, 2002]. During these, the EqL model values and the LTL and LTE model average values (and minima when they are reliable) show changes as large as those in MLS, consistent with previous studies showing that dynamical processes alone may account for the decrease in column ozone during minihole events [e.g., *Hood et al.*, 2001; *Allen and Nakamura*, 2002, and references therein]. Because minihole formation is accompanied by lifting of the tropopause and cooling of the lower stratosphere, PSCs often form in the same region as miniholes [e.g., *Hood et al.*, 2001; *Teitelbaum et al.*, 2001]; while this might be expected to induce more rapid ozone loss, *Hood et al.* [2001] have shown that heterogeneous processing during these events is not rapid enough to have a significant effect on column ozone values. The ability of several dynamical models to reproduce changes seen during these events demonstrates the dominance of dynamical processes in minihole formation, and is consistent with the expectation of no more than minor effects of enhanced chemical loss.

Minima in the EqL model on 20-21 February 1996 are ∼30 DU higher than MLS, and ∼25-40 DU higher on 29 February–3 March; LTE minima are ∼30 DU higher than MLS on 29 February–3 March. Qualitatively similar results are obtained for other (although less dramatic) minihole events during the MLS record. That the LTE and EqL model results give similar values suggests that interannual variability in large-scale transport does not play an important role in the dynamically produced interannual variability (since the EqL model does not include that process, and the climatological fields used in it are an average that would not be expected to closely reproduce the values for any particular year, large interannual variability in large-scale transport would be expected to result in significantly different values in the LTE and EqL models).

Because of the frequent lack of spatial correlation of column ozone with the lower stratospheric vortex (e.g., Figure 1, and section 2), the range of Col100 values (minimum to maximum) within the vortex is very large, in some years over 200 DU (e.g., 1996, black curves). Each of the dynamical models shows as much interannual variability as MLS in the maximum, and nearly as much in the minimum. However, the dynamical models show only ∼1/2 the variability in the average over most of the period shown (∼3/4 near the beginning, when less chemical ozone loss is expected to have already occurred), suggesting a substantial contribution of chemical loss to interannual variability. An increasing trend in LTL and LTE Col100 values with respect to MLS (except in the warm 1998 late winter) indicates ongoing chemical loss.

Interannual variability in vortex ozone averaged over 14-28 February from MLS and each dynamical model is shown in Figure 5, along with 46-hPa temperatures averaged over the same area and time period (these do not always reflect the overall relative "coldness" of the winters, but do indicate the type of variability that would be expected to arise at this time from adiabatic motions; i.e., lower Col100 values are expected during colder periods). This averaging period is near the beginning of the MLS late winter north-viewing periods and thus near the start of the LTL runs (green lines) in the first few years (see Figure 4), so the LTL values for 1992 and 1993 are still close to those from MLS. EqL model results calculated using NCEP/CPC data show differences of up to ∼10 DU (slightly smaller for minimum values), but the same amount of interannual variability. The close correlation of the EqL model results with temperature reflects the strong dependence of that model on the motion of the isentropes.

The difference between the LTE (blue) and MLS (black) Col100 values gives an estimate of chemical loss in the vortex averaged over 14-28 February. The amount of chemical loss estimated for the minimum, average, and maximum in 1996 (the year with most ozone loss, and a time with repeated minihole events) is ∼25, 50, and 70 DU, respectively. In the cold regions, especially when miniholes occur, the isentropes are raised and spread out, while in warm regions they are lowered and compressed. Since the ozone mixing ratio is approximately constant following a PV contour on an isentropic surface (i.e., at a particular location with respect to the vortex), and increases with increasing $\theta$ in the lower stratosphere, this results in lower mixing ratios at a given pressure, and hence lower Col100, in the cold regions than in warm regions, as was seen in Figures 1-3. Since chemical loss also follows PV contours on an isentropic surface (because loss occurring in one regions is rapidly transported around the vortex along PV contours), in the warm regions the mixing ratio decrease along a PV contour will be at lower pressure where is contributes more to the column. The difference seen in Figure 5 between estimates of column loss in minimum, average, and maximum Col100 arises from this effect; similar differences in loss estimated from minima, averages and maxima are seen in other years with large loss, especially 1993, which also had strong minihole activity. This effect may be amplified if column loss is estimated from loss calculated in an isentropic layer (as is done in many studies), as those calculations may exclude changes underneath the lowest isentropic layer in the cold region. This type of effect has important implications for estimating column ozone loss in the Arctic where both the vortex and the lower stratospheric temperature fields are highly asymmetric, especially from datasets with sparse or nonuniform coverage – if sampling is concentrated in warm (cold) regions, then column ozone loss may be over (under)-estimated, and the

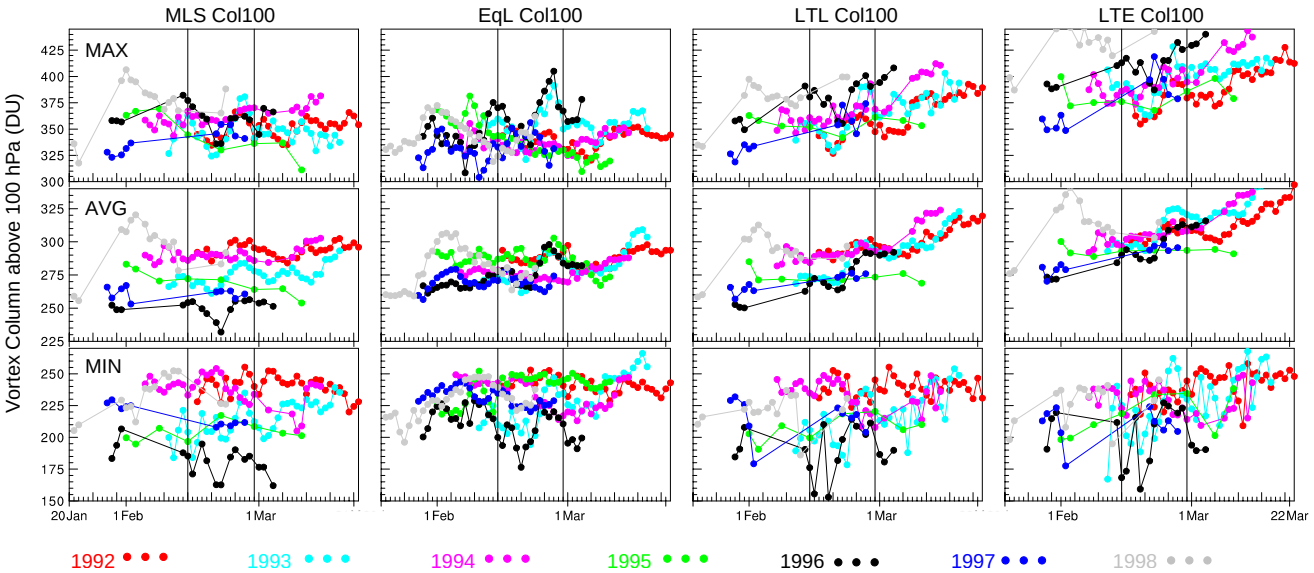


**Figure 4.** (Top to bottom) Maximum, average, and minimum daily Col100 in the 465-K vortex region from (left to right) MLS, EqL model, LTL runs, and LTE runs, during late winter 1992-1998. Thin vertical lines demark the 14-28 February period of the averages shown in Figures 1-3 and 5.

amount of that bias will vary interannually with the relative positions of the vortex, cold region, and sampling.

Interannual variability in minimum vortex Col100 is ∼60 DU for MLS, and ∼40-50 DU for the dynamical models, so the dynamical processes can account for 70-80% of the interannual variability in the minimum. Interannual variability in the maximum is ∼40 DU, 40 DU, 50 DU and 60 DU for MLS, EqL model, LTL runs and LTE runs, respectively; hence, dynamical processes may account for all variability in the maximum. In the average, however, the MLS data show ∼40 DU difference between the years, and the dynamical models ∼20 DU. Interannual variability in chemical ozone loss thus accounts for about half of the interannual variability seen by MLS in vortex-averaged Col100 in late February. When similar calculations are done for a latitude-band average over 64-80°N (similar to the average shown by *Andersen and Knudsen* [2002], but limited by the poleward extent of MLS observations, not shown), very similar conclusions are reached regarding the relative roles of dynamical and chemical variability, although the amount of variability is larger (∼60 DU in MLS and ∼30 DU in dynamical models). The EqL model shows about 2/3 to nearly all of the amount of variability seen in the LTE runs; this indicates that a large fraction of the interannual variability in dynamical processes is not, as postulated by *Chipperfield and Jones* [1999], directly related to variability in descent and large-scale transport, but rather is related to interannual variability in short timescale processes involving adiabatic warming/cooling and advection of subtropical low-ozone air into high latitudes, i.e., those processes primarily responsible for day-to-day variations in column ozone.

Previous estimates [*Chipperfield and Jones*, 1999; *Andersen and Knudsen*, 2002] of interannual variability in high latitude column ozone were for March averages. While MLS only observed the Arctic in most of March during its first two years of operation, we can estimate the minimum interannual variability related to dynamical effects using the EqL model, which depends only on having PV derived from meteorological analyses to reconstruct the 3D ozone fields. Figure 6 shows vortex-averaged EqL Col100 for 1992-2001 calculated using PV from Met Office data, and for 1979-2001 calculated using NCEP/CPC data, along with 46-hPa temperatures averaged for the same region and period. Comparing the values for 1992-1998 with those shown in Figure 5 for 14-28 February, we see a similar amount of interannual variability in the March average Col100 as in the late February average, about 20 DU in each period. Thus we expect about the same amount of interannual variability in March as in late February arising from the dynamical processes included in the EqL model (adiabatic motion of the isentropes, isentropic advection into high latitudes) and the changes in the area of the lower stratospheric vortex (the averaging area). Figure 6 indicates that the 7-year period observed by MLS was in fact a period of unusually small interannual variability in these dynamical processes. 1990-1998 was also a period of lower interannual variability in temper-

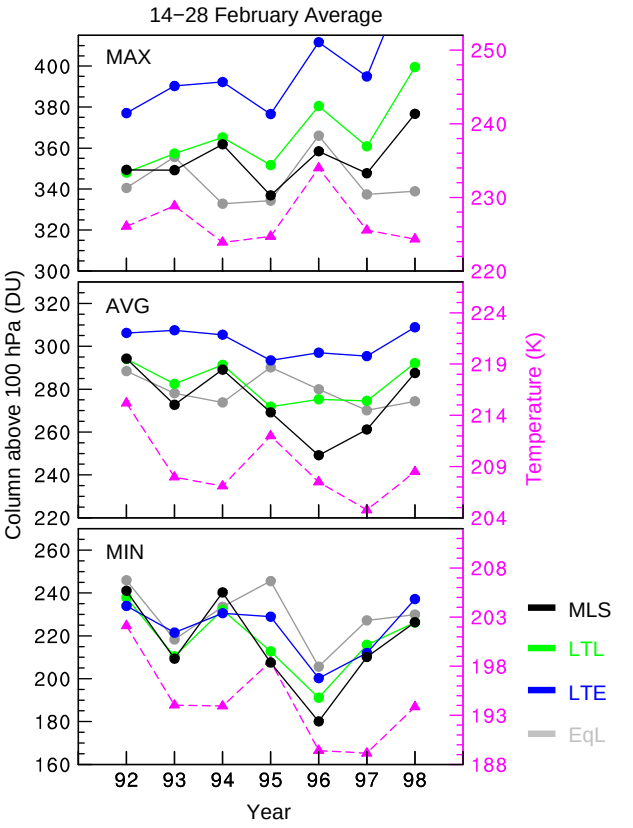


**Figure 5.** 14-28 February averages of daily (top to bottom) maximum, average, and minimum Col100 in the 465-K vortex, for (black) MLS data, (grey) EqL model, (green) LTL runs, and (blue) LTE runs. Magenta line and triangles show Met Office 46-hPa temperatures.

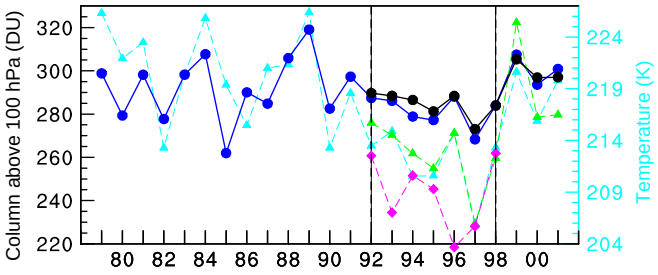


**Figure 6.** March averages of Col100 in the 465-K vortex region calculated from the EqL dynamical model, using PV from Met Office data (black) for 1992 through 2001, and from NCEP/CPC data (blue) for 1979 through 2001. Green and cyan lines and triangles show 46-hPa temperatures averaged over the same area and time period from Met Office and NCEP/CPC data, respectively. Magenta line and diamonds show estimates of column ozone in March accounting for MLS-observed chemical loss (see text). Thin vertical lines demark years observed by MLS.

ature; no major stratospheric sudden warmings, which often take place in late winter and strongly affect the late winter and spring vortex, occurred during this period, in contrast to an occurrence frequency of about one every two years before 1992, and more often after 1998 (a total of five in three of the four following winters) [e.g., *Naujokat et al.*, 2002, and references therein].

The dynamical processes in the EqL model result in only slightly lower average Col100 over 1992-1998 than over the earlier years, suggesting that these processes could account for only a very small fraction of observed lower column ozone values. Although variability in large-scale seasonal transport might also be expected to lower the values in colder years, our results for late February indicated that interannual variability in large-scale transport was a relatively small factor in changing Col100 values. Thus, the similar average values in the EqL model in the 1980s and 1990s suggest that chemical loss was an important factor in producing low values in the 1990s. *Manney et al.* [2002a], and references therein, have shown that only in 1996 and 1997 is chemical ozone loss expected to have continued significantly after the latest date observed by MLS in the winter. For 1996 and 1997 they estimated a maximum total loss by assuming loss continued at the late February rate through March. Using these values, and the total loss estimates for other years given by *Manney et al.* [2002a], March average loss in vortex Col100 is expected to be about 29, 54, 35, 36, 75, 45, and 22 DU in 1992, 1993, 1994, 1995, 1996, 1997, and 1998, respectively. Estimates of chemical loss from MLS are generally lower than those from other methods and datasets, but

within the combined uncertainties [e.g., *Harris et al.*, 2002; *Manney et al.*, 2002a], so these values represent a conservative estimate of the amount of chemical loss. Although the uncertainties in all such ozone loss estimates are large ($\sim$20%), we may reasonably expect that model biases are consistent from year to year because our estimates are obtained from a single dataset with consistent coverage using the LT model in the same way for all cases; they thus are expected to provide a good estimate of the interannual variability in chemical loss even though the uncertainties in the magnitudes of the loss are large,

The magenta symbols in Figure 6 show the expected Col100 values obtained by subtracting the estimates given above from MLS from the EqL model values, and suggest that total variability in vortex Col100 for March might be $\sim$45 DU in 1992-1998, over twice that in the EqL model. Interannual variability in descent/large-scale transport, which is not reflected in these estimates, would be expected to alter both EqL Col100 and Col100 estimated from chemical loss in a similar manner, and thus have a small impact on the relative amounts of interannual variability. The difference between a 1992-1998 average of the values estimated from MLS chemical loss and the EqL model average for 1979-1990 is close to the percent difference shown by *Andersen and Knudsen* [2002] for total column estimates ($\sim$14-17%, their Figure 1, an update of *Newman et al.* [1997]'s Figure 1). The variability in these estimates from MLS is qualitatively very similar to that seen in the total column observations discussed by *Newman et al.* [1997] and *Andersen and Knudsen* [2002], except that the estimate based on MLS is lower for 1996 (when the total column is not from TOMS as it is in the other years, and when the vortex and lowest column ozone region often extended equatorward of 63°N, the low latitude boundary of the averaging region for the total ozone estimates) relative to the other years. While these estimates are crude, and rely on several assumptions, they confirm that we would expect similar results for March as for late February regarding the relative roles of chemical and dynamical processes in interannual variability of Col100, and that chemical loss can explain much of the observed lower column ozone values in the 1990s.

Since MLS data only extend down to 100 hPa, our quantitative results discussed above apply directly only to Col100, not to total column. An estimate of the variability arising from short timescale dynamical processes in a more complete column is obtained by running the EqL model to reconstruct column ozone above 215 hPa (Col215), using the same EqL/$\theta$ climatology based on MLS data, but with values at levels below 100 hPa based on a zonal mean a priori field (from NASA/Langley). Col215 contains most ($\gtrsim$95%) of the total column at high latitudes, and all of the levels where chemical ozone loss occurs [e.g. *Goutail et al.*, 1999, 2001]. Col215 from the EqL model for both 14-28 February and March averages shows $\sim$25 DU interannual variability in the 1990s, the same percent of the actual values ($\sim$7%) as does Col100. *Goutail et al.* [1999, 2001] and *European Commission* [2001] show results suggesting that interannual variability in chemical loss below 100 hPa also follows similar patterns to that at higher levels. Thus, while we cannot quantify the relative contributions below 100 hPa, we have no reason to expect significantly different behavior in total column ozone.

The MLS instrument to be flown on the Earth Observing System Aura satellite scheduled for launch in 2004 will offer both continuous daily coverage of Arctic and Antarctic polar regions and ozone profiles extending down to $\sim$6 km. These data, and coincident $N_2O$ data (also measured by MLS on Aura) for use in verifying transport calculations, can provide much more precise estimates of the origins of variability in column ozone. When Aura MLS observations are combined with complementary measurements of tropospheric ozone from the Tropospheric Emission Spectrometer, column ozone from the Ozone Monitoring Instrument, and stratospheric ozone and long-lived tracer data from the High Resolution Dynamics Limb Sounder, the Aura platform will provide very powerful tools for understanding in detail the structure of ozone throughout the atmosphere and the effects of various chemical and dynamical processes on the evolution of total ozone.

## 4. Summary

MLS column ozone above 100 hPa (Col100) is examined in the NH late winters 1992-1998 to assess the relative effects of chemical and dynamical processes on its variability. Comparison of Col100 calculated using dynamical models with MLS Col100 shows that about half of the interannual variability in stratospheric column ozone averaged in the lower stratospheric vortex in February/early March is related to interannual variability in chemical loss; this is consistent with the analysis of total column ozone by *Andersen and Knudsen* [2002] using different methods and datasets, and confirms the prominent role of vortex chemical ozone loss in producing both larger interannual variability and lower column ozone in the 1990s than in the 1980s. The majority of the remaining interannual variability in Col100 (when that in chemical loss is taken out) is related to variability in the adiabatic motion of the isentropes and in short timescale quasi-isentropic transport associated with dynamical variations, rather than to interannual variability in descent rates and large-scale seasonal transport as was suggested by *Chipperfield and Jones* [1999].

Day-to-day variability in individual Arctic winters is of similar magnitude in MLS data and the dynamical models, and tracks closely in time, indicating the dominance of dynamical effects in producing short-term changes in Col100. Even during the Arctic winters with the greatest chemical ozone loss, and especially during winters when Col100 is poorly correlated with the lower stratospheric vortex, the morphology of Col100 in the MLS data is very similar to that in the purely dynamical models. Since chemical loss takes place inside the vortex and the patterns of chemical loss are closely correlated with the vortex, this implies that dynamical processes are still dominant in controlling the morphology of column ozone; this is in contrast to the southern hemisphere, where vortex chemical loss is so large that column ozone is always strongly correlated with the lower stratospheric vortex in late winter and spring.

Dynamical model calculations for 1979 through 2001 indicate that 1992-1998 was a period of less-than-usual interannual variability in late winter/spring temperature and in variations in Col100 related to short timescale dynamical processes; this result is also valid for total column calculated from the dynamical model. These dynamical processes could account for little of the lower observed total column ozone during the period, indicating that chemical loss was important in producing the low column ozone values observed in the 1990s. Our results indicate that, despite the dominance of dynamical processes in controlling day-to-day evolution and morphology of Arctic column ozone, chemical ozone loss, and interannual variability therein, were major contributors to both the large interannual variability in and lower values of late winter/spring column ozone in the 1990s.

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